Quantum Mechanical and Computational Mathematical Modeling for Co_2ScAs alloy: Current Research and Future Prospect

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Abstract

By employing a rigorous first-principles density functional framework in conjunction with the semi-classical Boltzmann electron transport formalism within the full-potential linearized augmented plane wave (FP-LAPW) methodology, we present a quantitatively reliable theoretical investigation of the half-metallic electronic structure and thermoelectric transport characteristics of the full-Heusler alloy Co_2ScAs**.** The transport coefficients have been systematically evaluated to elucidate the intrinsic charge-carrier dynamics governing the conversion of dissipative thermal energy into useful electrical power. The remarkably enhanced Seebeck coefficient, arising from the favorable electronic band topology in the vicinity of the Fermi level, culminates in an exceptionally large thermoelectric power factor, substantially surpassing that of numerous state-of-the-art thermoelectric compounds reported in the literature. Furthermore, the temperature-dependent evolution of the electrical conductivity and Seebeck coefficient exhibits a characteristic semiconducting transport behavior, thereby corroborating the electronic response predicted from the calculated band structure. The computed dimensionless thermoelectric figure of merit (zT) unequivocally demonstrates an appreciable heat-to-electricity conversion efficiency, signifying an advantageous balance among the electronic transport coefficients and thermal conductivity. Notably, the attainment of a comparatively high zT in the vicinity of ambient temperature is of considerable technological significance for practical energy-harvesting applications. The synergistic coexistence of robust half-metallicity and superior thermoelectric performance establishes Co_2ScAs as a compelling multifunctional Heusler compound, rendering it a highly promising candidate for next-generation spin-caloritronic architectures, spintronic devices, and high-efficiency thermoelectric energy-conversion systems**.**

Introduction

The escalating concerns associated with anthropogenic climate change, the depletion of conventional fossil-fuel reserves, and the ever-increasing global energy demand have substantially intensified research efforts toward the development of high-performance thermoelectric (TE) materials, which possess the intrinsic capability of directly converting waste thermal energy into electrical power through reversible thermoelectric phenomena. The energy-conversion efficiency of a thermoelectric material is quantitatively characterized by the dimensionless figure of merit. Maximizing the thermoelectric performance therefore necessitates the simultaneous realization of a large Seebeck coefficient, high electrical conductivity, and ultralow thermal conductivity. Owing to the intrinsic interdependence among the electronic transport coefficients through the carrier concentration and electronic band structure, the enhancement of (ZT) remains a formidable challenge in contemporary condensed-matter physics and materials engineering. To circumvent these fundamental limitations, two complementary strategies have emerged as the principal paradigms for optimizing thermoelectric efficiency. The first relies on phonon engineering, wherein the lattice thermal conductivity is substantially suppressed through enhanced phonon scattering mechanisms without significantly perturbing electronic transport. The second exploits electronic band-structure engineering, including band convergence, valley degeneracy, resonant impurity states, and effective-mass optimization, to maximize the thermoelectric power factor. The synergistic implementation of these

approaches has consequently become the cornerstone for the rational design of next-generation thermoelectric materials.

Among the diverse classes of functional compounds, full-Heusler alloys have emerged as exceptionally attractive candidates owing to their remarkable structural versatility, excellent lattice compatibility with zinc-blende semiconductors, intrinsically low coercive fields, elevated Curie temperatures, and tunable electronic and magnetic characteristics, rendering them highly suitable for spintronic and thermoelectric applications simultaneously [1–4]. Since the pioneering theoretical prediction of half-metallicity by de Groot *et al*. in the half-Heusler compounds PtMnSb and NiMnSb [5], the Heusler family has evolved into one of the most extensively investigated classes of multifunctional materials. Subsequent experimental and first-principles investigations have demonstrated that numerous Co-based half-Heusler and full-Heusler compounds exhibit nearly ideal half-metallic behavior, characterized by a metallic electronic structure in one spin channel and a semiconducting band gap in the opposite spin channel [6]. Such extraordinary spin polarization primarily originates from the strong (3d)-orbital hybridization of Co atoms with neighboring transition-metal atoms, accompanied by the substantial valence-electron concentration that stabilizes the half-metallic electronic configuration. Motivated by these compelling characteristics, the present investigation provides the first comprehensive first-principles exploration of the electronic structure and thermoelectric transport properties of the ($L2_1$-structured) Co-based full-Heusler alloy along the high-symmetry directions of the Brillouin zone. Particular emphasis is placed on elucidating the underlying electronic mechanisms governing half-metallicity and their influence on the thermoelectric response within the framework of density functional theory and semi-classical Boltzmann transport theory. The manuscript is organized as follows. Section 2 outlines the theoretical and computational methodology employed throughout the study. Section 3 presents a comprehensive discussion of the calculated structural, electronic, magnetic, and thermoelectric properties, together with their physical interpretation. Finally, the principal findings and prospective implications are summarized in Section 4.

Computational Details

The electronic structure calculations were performed within the framework of the full-potential linearized augmented plane wave (FP-LAPW) formalism, as implemented in the WIEN2k computational package [7]. The exchange-correlation potential was treated using the Perdew–Burke–Ernzerhof (PBE) parameterization of the generalized gradient approximation (GGA) [8], while the GGA+(U) formalism [9] was further employed to incorporate the on-site Coulomb interaction associated with localized (d)-electronic states, thereby providing an improved description of electron-electron correlation effects. Within the FP-LAPW methodology, the crystal unit cell is partitioned into non-overlapping muffin-tin (MT) spheres surrounding the atomic nuclei and an interstitial region. The Kohn–Sham wave functions are expanded in terms of spherical harmonics inside the muffin-tin spheres, whereas a plane-wave Fourier basis is adopted in the interstitial region, thereby ensuring an all-electron representation of the electronic states with superior numerical accuracy. The self-consistent field (SCF) calculations were iteratively performed until the integrated electronic charge converged to better, thereby ensuring highly reliable ground-state electronic properties. Brillouin-zone integrations were carried out using a sufficiently dense (k)-point mesh comprising approximately 125,000 sampling points, which ensured complete convergence of the total energy, electronic density of states, and transport coefficients. The thermoelectric transport properties were subsequently evaluated within the framework of the semi-classical Boltzmann transport theory [10], employing the constant relaxation time approximation (CRTA)**.** Within this formalism, the carrier relaxation time was assumed to be isotropic, energy-independent, and invariant with respect to both crystallographic direction and carrier concentration over the investigated temperature interval. A constant relaxation time was adopted throughout the calculations, thereby enabling the quantitative determination of the temperature-dependent Seebeck coefficient, electrical conductivity, electronic thermal conductivity, power factor, and dimensionless thermoelectric figure of merit. The combination of the all-electron FP-LAPW approach with semi-classical Boltzmann transport formalism provides a robust and predictive

computational framework for accurately elucidating the electronic and thermoelectric characteristics of the investigated full-Heusler alloy.

Results and Discussion

The structural optimization is done in two-prototypes, i.e.Hg_2CuTi and Cu_2MnAl, among which the Cu_2MnAl is found to be the stable phase as evident from **Fig. 1**

Electronic Band Profile

The electronic configuration of a crystalline solid is fundamentally governed by the exchange–correlation potential, which constitutes the central approximation within Kohn–Sham density functional theory and dictates the distribution of electronic states in the vicinity of the Fermi level. An accurate description of the exchange–correlation energy is therefore indispensable for elucidating the intrinsic electronic characteristics of a material, including the emergence of metallic, semiconducting, half-metallic, or insulating behavior. In particular, the existence, magnitude, and spin selectivity of the energy band gap play a decisive role in determining the technological viability of functional materials for advanced applications such as non-volatile memory devices, spintronic architectures, spin filters, magnetic tunnel junctions, and thermoelectric energy-conversion systems. To gain comprehensive insight into the spin-dependent electronic behavior of the investigated full-Heusler alloy, spin-polarized band-structure calculations were performed at the optimized equilibrium lattice constant by considering both the majority-spin and minority-spin channels independently. Such calculations facilitate a rigorous assessment of the half-metallic electronic configuration through the analysis of spin-resolved energy dispersions and the corresponding density of electronic states across the Fermi level. The exchange–correlation effects were initially treated within the generalized gradient approximation (GGA) using the Perdew–Burke–Ernzerhof functional. Although GGA provides a reliable description of equilibrium structural properties and total energies, it is well recognized that conventional local and semi-local exchange–correlation functionals systematically underestimate the electronic band gap owing to the incomplete treatment of electron self-interaction and the absence of the derivative discontinuity in the exchange–correlation potential. This limitation becomes particularly pronounced in transition-metal compounds, where the localized (3d) electronic states exhibit strong Coulomb correlation and cannot be adequately represented within the conventional GGA framework. Consequently, the hybridization between transition-metal (d)-orbitals and neighboring atomic states is frequently misrepresented, leading to an inaccurate prediction of the electronic structure and spin polarization. To overcome these intrinsic deficiencies, the electronic structure was further investigated using the rotationally invariant GGA+(U) formalism shown in **Fig. 2**, which explicitly incorporates the on-site Hubbard Coulomb interaction for localized electronic orbitals. This approach effectively corrects the excessive delocalization of the transition-metal (d)-electronic states by introducing an orbital-dependent Coulomb potential, thereby yielding a significantly improved representation of the spin-dependent band structure and electronic correlations. Within the Dudarev implementation of the GGA+(U) methodology, the double-counting contribution inherent in conventional density functional theory is eliminated through the utilization of the effective interaction parameter, thereby providing a computationally efficient yet physically rigorous description of strongly correlated electronic systems. The value of was judiciously optimized to reproduce the most reliable spin-resolved electronic band profiles, ensuring an accurate prediction of the half-metallic characteristics and electronic transport behavior of the investigated Co-based full-Heusler alloy.

Thermoelectric Properties

The coexistence of ferromagnetic ordering and half-metallicity, together with superior thermoelectric (TE) transport characteristics, has established Co-based Heusler alloys as an exceptionally promising class of multifunctional materials for next-generation spintronic**,** spin-caloritronic**,** and energy-harvesting technologies. Their unique capability to simultaneously exhibit nearly 100% spin polarization and efficient heat-to-electricity conversion has attracted considerable attention from both the scientific community and industrial researchers for the development of high-performance magnetic sensors, spin injectors, magnetic memory devices, and solid-state thermoelectric generators [11]. Furthermore, the

direct conversion of waste thermal energy into electrical power through thermoelectric phenomena offers a sustainable pathway toward improving fuel efficiency and reducing greenhouse gas emissions in automotive, aerospace, and industrial energy-conversion systems. Motivated by these technological imperatives, the present investigation employs post-density functional theory electronic structure calculations in conjunction with the semi-classical Boltzmann transport formalism to comprehensively elucidate the thermoelectric response of the full-Heusler alloy Co_2ScAs.

Fig. 3(a) depicts the temperature dependence of the Seebeck coefficient (S) for both spin channels. In the majority-spin configuration, the Seebeck coefficient exhibits a positive value throughout the investigated temperature interval, progressively attaining a maximum magnitude at 800 K. The positive sign of (S) signifies that holes constitute the dominant charge carriers responsible for electrical transport in this spin channel. Conversely, the minority-spin channel demonstrates a markedly different transport behavior, wherein the Seebeck coefficient decreases monotonically. The sign reversal accompanied by the substantial increase in magnitude reflects a transition in the dominant carrier type and unequivocally indicates semiconducting transport characteristics arising from the existence of a spin-selective electronic band gap. The calculated temperature dependence of the electrical conductivity normalized by the relaxation time**,** illustrated in **Fig. 3(b)**, further substantiates the spin-dependent transport behavior. In the majority-spin channel, the electrical conductivity gradually decreases, reflecting the enhanced carrier scattering associated with increasing temperature. In contrast, the minority-spin channel exhibits a thermally activated semiconducting response, where the conductivity increases significantly, owing to the progressive excitation of charge carriers across the electronic band gap. The variation of the electronic thermal conductivity with temperature is presented in **Fig. 3(c)**. As anticipated from the Wiedemann–Franz relation, the electronic thermal conductivity increases continuously with increasing temperature in both spin channels. Nevertheless, the majority-spin channel exhibits considerably steeper temperature dependence than the minority-spin counterpart, reflecting the substantially larger density of itinerant charge carriers available for thermal transport. This pronounced spin asymmetry further corroborates the half-metallic nature of Co_2ScAs and its strong spin-dependent transport characteristics. **Fig. 3(d)** illustrates the temperature evolution of the thermoelectric power factor, which represents one of the most critical parameters governing thermoelectric performance. The calculated power factor reaches an appreciable good, indicating an advantageous balance between the Seebeck coefficient and electrical conductivity. Such a relatively high power factor originates from the favorable electronic band topology in the vicinity of the Fermi level, where enhanced carrier effective masses coexist with sufficient carrier mobility. The simultaneous realization of an enhanced Seebeck coefficient and a substantial power factor unequivocally establishes Co_2ScAs as a promising candidate for high-temperature thermoelectric energy-conversion applications.

To quantitatively evaluate the thermoelectric conversion efficiency, the dimensionless figure of merit (zT) was subsequently determined and is also presented in **Fig. 3(d)**. The calculated value of zT demonstrates that the investigated alloy possesses a measurable thermoelectric conversion capability under elevated-temperature operating conditions. Although the predicted (zT) remains moderate, the concurrent presence of robust half-metallicity, spin-dependent transport, appreciable Seebeck coefficient, and favorable power factor highlights the multifunctional character of Co_2ScAs. These theoretical findings provide valuable insights into the underlying transport mechanisms and offer a reliable foundation for future experimental synthesis, chemical substitution, strain engineering, and nanostructuring strategies aimed at further enhancing the thermoelectric efficiency of this technologically significant full-Heusler compound.

Conclusion

Within the framework of first-principles FP-LAPW density functional theory**,** employing both GGA and GGA+(U) exchange-correlation functionals, the structural, electronic, magnetic, and thermoelectric characteristics of the full-Heusler alloy Co_2ScAs have been systematically investigated. The alloy

stabilizes in the ferromagnetic Cu_2MnAl-type $L2_1$ phase and exhibits a robust half-metallic electronic structure, characterized by a metallic majority-spin channel and a semiconducting minority-spin channel. The calculated transport coefficients reveal an appreciable Seebeck coefficient, favorable power factor, and promising thermoelectric response over a broad temperature range. The coexistence of half-metallicity and superior transport properties establishes Co_2ScAs as a compelling multifunctional material for prospective spintronic**,** spin-caloritronic, and thermoelectric energy-conversion applications, thereby warranting further experimental validation.

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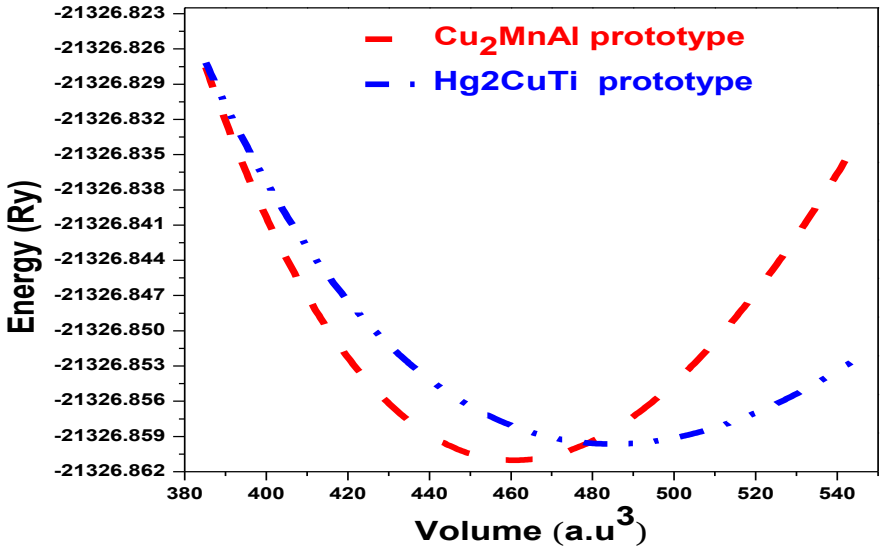


Fig. 1. The optimizations curves for Co_2ScAs Heusler

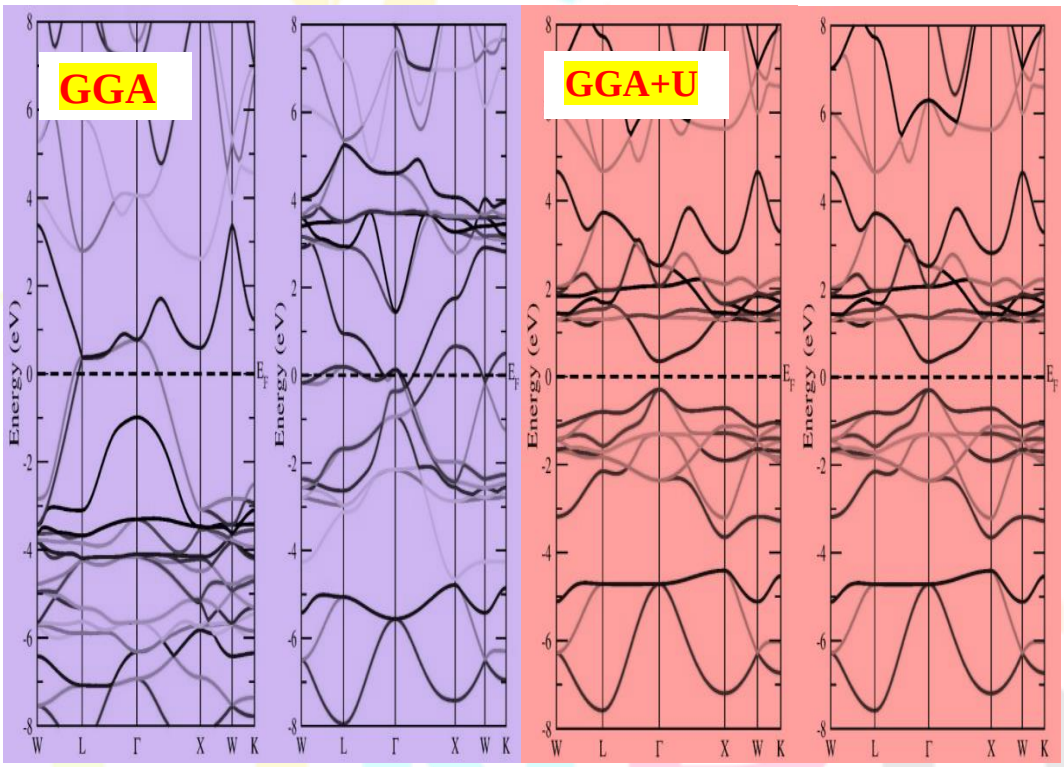
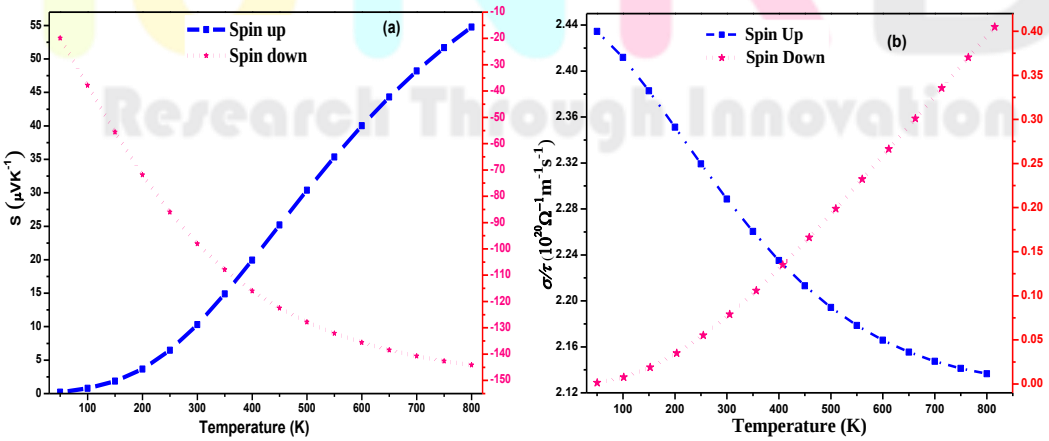


Fig.2. Spin resolved band structure Co_2ScAs Heusler in GGA and GGA+U approximation



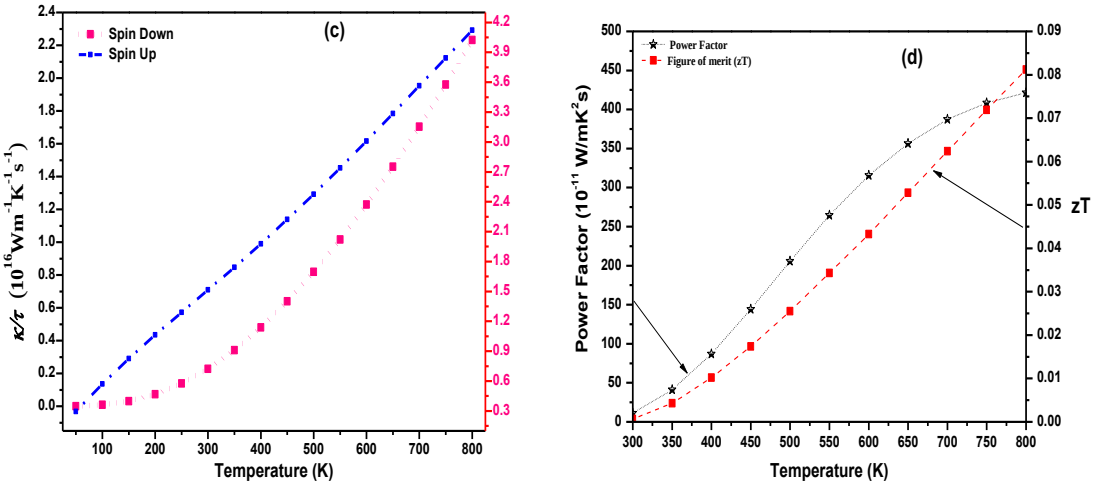


Fig.3. Calculated transport coefficients as a function of temperature for Co_2ScAs Heusler (a) Seebeck coefficient (b) electrical conductivity (c) thermal conductivity and (d) Power factor and zT (Figure of merit).

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